ATLAS PROJECT	Activation and activation-retention in a liquid argon calorimeter		
ATLAS Project Document No.: **ATL-A-TR-0003**	Institute Document No.:	Created: September 24, 2004	Page: **1 of 10**
		Modified: February 16, 2005 13:51	Rev. No.: **2**

Released for Information

Activation and activation-retention in a liquid argon calorimeter

Abstract

A small quantity of liquid argon in an aluminum cell was exposed to 1.55×10^{15} protons at 24 GeV/c in about an hour. Measurable quantities of the gamma-emitting isotopes ^{7}Be, ^{22}Na, ^{24}Na, ^{28}Mg, ^{38}S, ^{38}Cl, and ^{39}Cl were produced. About 50% of the measured ^{7}Be and ^{22}Na vented out of the cell into a pressure cylinder with the argon, while it was still in the liquid state. Within our ability to measure it, less than 1% of the ^{7}Be and ^{22}Na vented out with the gaseous argon from the pressure vessel. The other isotopes probably behaved similarly, but because of short half-lives or low activity, we were unable to detect all of the observed isotopes in all of the constituent samples. While some activity in the form of ^{38}Cl and ^{39}Cl did escape from the open, LAr bath used to keep the cell cold, it did not travel more than a few meters from the bath before sinking to the floor, where it remained until it decayed a few hours later.

Prepared by:

J. Cumming, D. Fournier, A. Gonidec, G. Greene, R. W. Hackenburg, A. Hanson, V. Hedberg, R. Holroyd, J. Y. Hostachy, D. Lissauer, L. Mausner, M. Nessi, H. Oberlack, C. Oram, V. Radeka, D. Rahm, and W. Willis

Checked by:

Approved by:

Distribution List:

1 Introduction

Both the endcap and the barrel calorimeters in the ATLAS experiment at the LHC [1] contain liquid argon (LAr), which will become activated when the LHC runs. The activation process was studied by Waters and Wilson in 1994 [2]. If a leak in a LAr calorimeter occurs while the argon is activated, gas will be released to the atmosphere with some level of radioactive contamination. In order to know how best to deal with these radioactive isotopes, the quantity of activity which would vent-out with the argon must be determined. Thus, our goals are to estimate the spallation cross sections for the production of radioactive isotopes from ^{nat}Ar with half-lives of about 30 minutes or more, and to determine the fraction and makeup of the isotopes retained in the calorimeter vessel, and the fraction that comes out with the argon when the vessel is emptied. We limit our study to the γ-emitting radioactive isotopes ^{7}Be, ^{22}Na, ^{24}Na, ^{28}Mg, ^{38}S, ^{38}Cl, and ^{39}Cl. These are fairly easy to measure and are representative of those which we did not measure.

2 Experimental Apparatus

We fabricated a test cell similar geometrically and electrically to the ATLAS calorimeters, along with a LAr bath to keep the cell cold (Figs. 1 and 2). The cell, also refered to as "the calorimeter", is connected to a pressure cylinder to hold the argon after exposure. The aluminum test cell holds about 22 cc of LAr. It contains electrodes, which are 2 mm thick discs of Lexan polycarbonate with 25 micron foils of Cu and stainless steel glued to the Lexan. The electrode structure has four gaps of 2 mm, filled with LAr, for a total LAr length of 8 mm. This construction, with the Lexan instead of solid Cu and stainless steel, was intended to minimize the amount of activity produced from materials other than the argon, while emulating the ATLAS calorimeter surfaces as closely as possible. Rohacel foam was used to exclude the LAr in the bath from direct exposure to the beam.

3 Exposure

We exposed the test cell to 24 GeV protons at the D line of the Alternating Gradient Synchrotron at Brookhaven National Laboratory. The total flux was 1.55×10^{15} protons, determined from beam counters calibrated to about 5%. 2 kV was applied to the cell before the exposure started, but the supply was unable to hold the voltage once the beam was turned on. Thus, the experiment was performed with no voltage on the electrodes.

4 Sample Preparation

After the exposure, the LAr in the cell was transfered to the evacuated aluminum pressure cylinder (chilled in LAr) under its own pressure by cryopumping. The cryopumping was accomplished by opening valves 3 and 4 (Fig. 2), then removing the LAr cell from the LAr bath and allowing it to warm to room temperature; the warming was accelerated by blowing hot air on the cell.

Bubbler water sample: The argon sat in the pressure cylinder at room temperature for about four hours while the apparatus was wrapped for transport, transported to a hot lab, unwrapped,

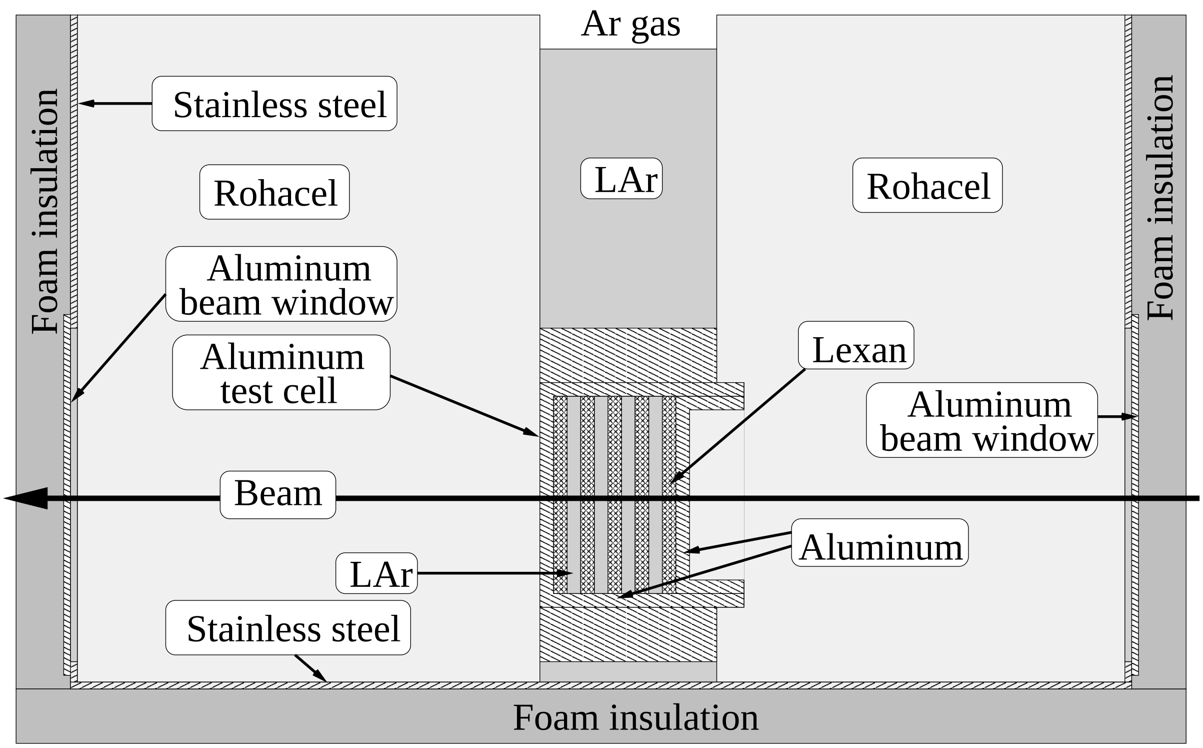

Figure 1: Side view of the apparatus used to expose LAr to a beam of protons while keeping it cold. The portion of the cell containing LAr is 4.74 cm in diameter. The total length of the LAr in the cell is 8 mm. The top is covered, but not hermetically. The sketch is not to scale.

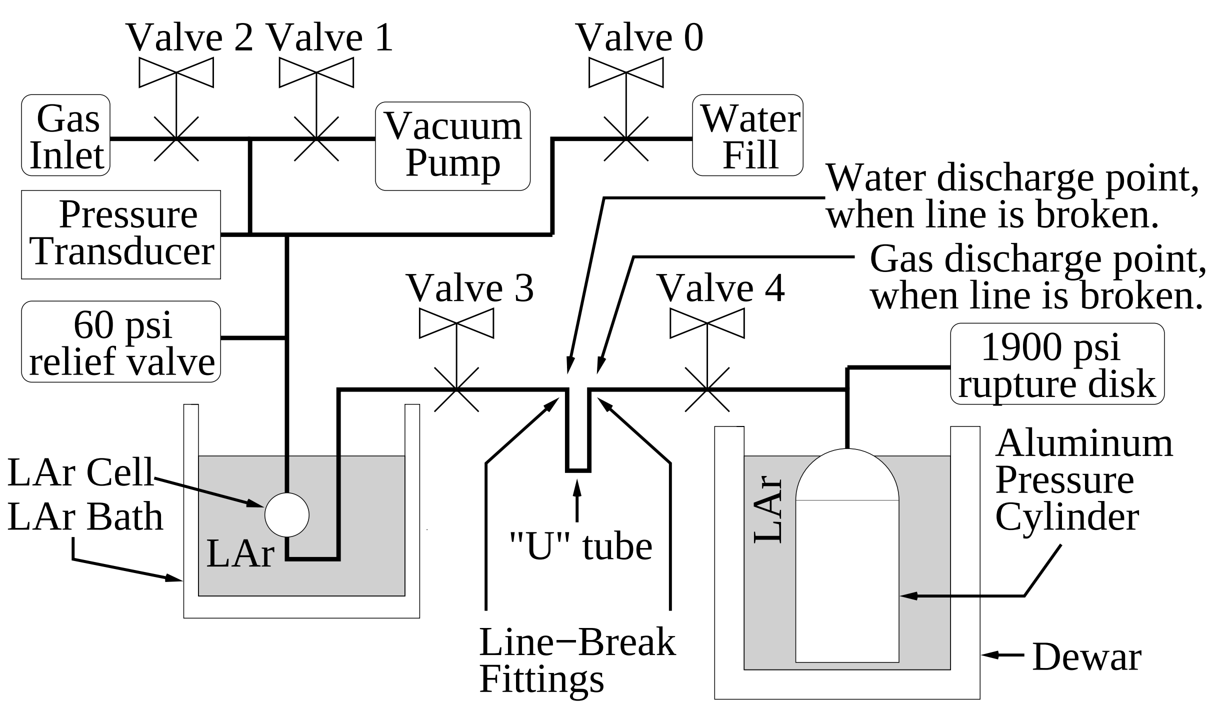

Figure 2: Schematic of the plumbing used to fill the cell and to transfer the exposed LAr to the pressure cylinder.

Table 1: Expected Isotope Production in Liquid Argon. Only those isotopes with half-lives greater than 30 minutes are shown. The decay data is from the Table of Isotopes [3].
σ_{MENDL}: Calculated and extrapolated cross sections from MENDL-2 [4], for 2.0 GeV protons.
σ_{WEBBER}: Measured cross sections from Webber [8], from 600 MeV/nucleon ^{40}Ar on hydrogen.

Isotope	Decay	Product	$t_{1/2}$	σ_{MENDL} (mb)	σ_{WEBBER} (mb)
^{3}H	β^-	^{3}He	12.3 y	50.3[a]	–
^{7}Be	EC,γ	^{7}Li	53.3 d	3.8[b]	–
^{10}Be	β^-	^{10}B	1.6 My	3.8	–
^{14}C	β^-	^{14}N	5.7 ky	3.8[b]	–
^{18}F	EC,β^+	^{18}O	110 m	3.8[b]	–
^{22}Na	EC,β^+,γ	^{22}Ne	2.60 y	3.4	–
^{24}Na	β^-,γ	^{24}Mg	15.0 h	5.8	–
^{26}Al	EC,β^+,γ	^{26}Mg	740 ky	6.0	2.2
^{28}Mg	β^-,γ	^{28}Al	20.9 h	0.086	–
^{31}Si	β^-	^{31}P	157 m	2.6	14.1
^{32}Si	β^-	^{32}P	172 y	–	3.4
^{32}P	β^-	^{32}S	14 d	20.7	28.1
^{33}P	β^-	^{33}S	25 d	8.47	18.9
^{35}S	β^-	^{35}Cl	88 d	26.6	26.0
^{36}Cl	β^-	^{36}Ar	300 ky	11.3	26.3
^{37}Ar	EC	^{37}Cl	35 d	–	5.9
^{38}S	β^-,γ	^{38}Cl	170 m	0.006	0.4
^{38}Cl	β^-	^{38}Ar	37.2 m	–	25.1
^{39}Cl	β^-,γ	^{39}Ar	55.6 m	5.4	28.8
^{39}Ar	β^-	^{39}K	269 y	–	60.1

[a] The cross section for ^{3}H is for aluminum, as ^{27}Al, instead of argon.
[b] This is the ^{10}Be cross section.

and finally connected to a pair of conventional bubblers in series. The argon was then slowly passed through the bubblers at about 1.5 L/min, which took about 15 min. Thus, two bubbler water samples were produced, but the activity in the first sample was so low that the second sample was ignored; the water from the first bubbler is henceforth referred to as the "bubbler water".

Calorimeter rinse sample: An aqueous solution of HCl was used to flush the cell.

Cylinder rinse sample: The pressure cylinder was flushed with an aqueous solution of HCl.

Plumbing rinse sample: The plumbing was flushed with an aqueous solution of HCl.

Table 2: Contributions to the cross sections (in mb) from measured activity in the samples (see text). The "Bubbler water" contributions for ^{7}Be, ^{22}Na, and ^{28}Mg were only determined as upper limits because the activities were too small. These are comparable to the overall uncertainties in their respective apparent cross sections and are ignored in taking the sums, but are used to set upper limits for the fractions discharged with the argon gas. Entries with "No peak" are those for which no γ-peak was identified by the analysis software. The errors on the cross section contributions are standard deviations, propagated from the errors in detected activity determined by the analysis software. See text for explanations of "Estimated max.", F(gas), and F(LAr). "Expected" are the cross sections from Table 1. Uncertainties are standard deviations, shown in parenthesis as the error in the least significant digits.

	^{7}Be	^{22}Na	^{24}Na	^{28}Mg	^{38}S	^{38}Cl	^{39}Cl
Bubbler water	< 0.25	< 0.62	0.0111(11)	< 0.007	0.00273(46)	0.192(24)	0.0721(39)
Calorimeter	2.85(19)	2.61(35)	3.124(60)	0.617(16)	0.439(11)	21.93(52)	7.43(14)
Cylinder	1.94(17)	1.53(22)	No peak	No peak	No peak	No peak	No peak
Plumbing	1.02(21)	0.57(22)	No peak	No peak	No peak	No peak	No peak
Samples sum	5.81(41)	4.71(78)	3.135(60)	0.617(16)	0.442(11)	22.12(52)	7.50(14)
Estimated max.	5.81(41)	4.71(78)	5.7(1.2)	1.11(24)	0.80(17)	39.9(8.5)	13.5(2.9)
Expected	3.8	3.4	4	0.086	0.4	25.1	28.8
F(gas) (%)	< 4.3	< 13	0.196(46)	< 0.63	0.342(93)	0.48(12)	0.53(12)
F(LAr) (%)	50.9(5.9)	44.6(9.9)	45(23)	45(24)	45(24)	45(23)	45(23)

5 Analysis

Cross sections: Let A be the measured activity of isotope X, which has a characteristic decay rate λ, counted for a duration Δt at a time t following an exposure of a thin, uniform target of number-density n and thickness d to a flux Φ of duration T. The apparent cross section is

$$\sigma = A \frac{\Delta t e^{\lambda t}}{nd\Phi Q\left(1 - e^{-\lambda \Delta t}\right)}, \tag{1}$$

where Q is the saturation factor, typically

$$Q = \frac{1 - e^{-\lambda T}}{\lambda}. \tag{2}$$

An interrupted or time-dependent exposure may be handled through a modification to Q. If the activity is divided into a complete set of N samples $\{A_i\}$, with

$$A = \sum_{i=1}^{N} A_i, \tag{3}$$

then a cross section contribution σ_i may be obtained from A_i for each sample with (1). The sum of these contributions is the production cross section for isotope X,

$$\sigma = \sum_{i=1}^{N} \sigma_i. \tag{4}$$

The primary reaction for producing isotope X in a thin target with a pure beam is

$$p + {}^{40}\mathrm{Ar} \rightarrow \mathrm{X} + \mathrm{Y}, \tag{5}$$

where Y represents one or more nuclei or nucleons. The thin-target approximation neglects the removal of primary particles from the beam due to collisions with target particles upstream of other target particles. Neglecting this here results in a small fraction of a percent error. There is a further, very substantial error in neglecting the production of secondary particles in a thick target, which themselves may interact in the LAr. These make contributions such as

$$n + {}^{40}\mathrm{Ar} \rightarrow \mathrm{X} + \mathrm{Y}'. \tag{6}$$

Other materials in the target region contribute, through

$$p + \mathrm{A} \rightarrow \mathrm{X} + \mathrm{A}'. \tag{7}$$

However, this contribution is small because the recoil of X must be large enough to escape the material in which it is produced while small enough to be captured in the LAr.

Seven isotopes (^{7}Be, ^{22}Na, ^{24}Na, ^{28}Mg, ^{38}S, ^{38}Cl, and ^{39}Cl) were produced in measurable quantities. No measurement of ^{40}K activity was significantly above room background. None of the other detected isotopes had any measurable room background activity. Table 2 lists the cross section contributions from activity detected in each of the four constituent samples (bubbler water, calorimeter rinse, plumbing rinse, cylinder rinse). The bubbler water and calorimeter rinse samples were counted a second time, after a delay, to allow the shorter-lived isotopes to decay. This reduced the background enough to permit a determination of the activity of the longer-lived isotopes ^{22}Na and ^{7}Be in the calorimeter rinse. No activity was detected in the delayed bubbler water counting. For the few isotopes where activity was measured for both the calorimeter rinse and delayed calorimeter rinse countings, the former is the more precise and is the one used.

Activated samples were placed in the sample-space of a hyper-pure Ge (HPGe) detector [10] and γ-peak strengths were obtained. The analyzer software determines the activity (and uncertainties, as standard deviations) for each γ peak it recognizes from a library of peaks, corrected for detector efficiency, sample size, time delay since end-of-exposure, and branching ratios of the identified peaks. In principle, the four samples compose a complete set, and the total cross sections are given by the sum (4) of their contributions (1) calculated from their measured activities, thus

$$\sigma = \sigma(\text{Bubbler water}) + \sigma(\text{Calorimeter rinse}) + \sigma(\text{Cylinder rinse}) + \sigma(\text{Pumbing rinse}). \tag{8}$$

However, two of the samples (the cylinder rinse and the plumbing rinse) were not counted before a long delay (more than 12 days), because not much was expected there. As a result, those samples' contributions in (8) are not known for the isotopes ^{24}Na, ^{28}Mg, ^{38}S, ^{38}Cl, and ^{39}Cl, which have half-lives less than a day. For each isotope which had measureable activity in the cylinder and plumbing rinses (^{7}Be and ^{22}Na), the estimated maximum cross section is the sum (8).

Estimated maximum cross sections (reconstructed sums): To estimate the missing contributions for the cylinder and plumbing rinses for the isotopes ^{24}Na, ^{28}Mg, ^{38}S, ^{38}Cl, and ^{39}Cl, we make the reasonable assumption that the two isotopes of sodium are like-proportioned amongst the samples. From Table 2, for ^{22}Na,

$$\frac{\sigma(\text{Sample sum})}{\sigma(\text{Calorimeter rinse})} = 1.80(38). \tag{9}$$

This is the ratio of total activity to that which did not vent from the cell. The corresponding ratio from the ^{7}Be is 2.04(20), and the weighted average of the ^{7}Be and ^{22}Na ratios is 1.99(24). The ratios from ^{7}Be and ^{22}Na are not significantly different. The ratio 1.80(38) from ^{22}Na is used to obtain the reconstructed sums in Table 2 for the isotopes ^{24}Na, ^{28}Mg, ^{38}S, ^{38}Cl, and ^{39}Cl. That ratio is applied to the contributions for those isotopes from the calorimeter rinse sample to obtain the reconstructed sums. For example, for ^{24}Na,

$$\sigma(\text{Reconstructed sum}) = 1.80(38) \times \sigma(\text{Calorimeter rinse}) = 5.7(1.2)\ \text{mb}. \tag{10}$$

While this method of reconstructing the sums for different isotopes (using this same ratio) seems reasonable for the metals, and even for the sulfur, it is most suspect for the chlorine isotopes. Any chlorine which vented to the cylinder as a solute in the LAr or as a Cl_2 precipitate (solid at LAr temperatures) should have vented out of the cylinder with the gaseous argon. If the nascent chlorine formed any compounds at all, including Cl_2, it was most likely with contaminants in the LAr, such as oxygen or methane, which had much greater concentrations than the nascent chlorine, even though they were present at only a few parts per 10 million. That is, compounds such as methyl chloride were more likely to be formed than Cl_2. Nevertheless, any such compound that we could conceive of is a solid at LAr temperature, and a gas at room temperature. Assuming the ratio (9) is not too far off for the chlorine, a substantial fraction of the chlorine must have vented to the cylinder with the LAr, but then remained in the cylinder, possibly by attaching to the cylinder wall, when the argon gas was vented through the bubblers. It is quite possible that the chlorine attached to the cylinder walls with much greater efficiency than it did in the cell, because the cylinder sat for nearly 4 hours at room temperature before being vented through the bubblers, whereas the chlorine sat for an hour or less at LAr temperature before being transferred to the pressure cylinder.

The Aluminum foil monitor: A hyperpure Al foil was used to monitor the exposure. The measured cross section for production of ^{24}Na from Al is 8.78(14) mb for 22.4 GeV/c protons [9]. The ^{24}Na activity we observed in the Al foil sample corresponds to a cross section of 15.96(41) mb, through (1). This is a factor 1.818(55) too large for the total flux delivered. Since we had fairly good control over the beam intensity, it is not possible to explain the large activity as being due to a larger-than-expected flux. Contamination from ^{24}Na produced in the LAr bath is indicated (but not quantified), since ^{38}Cl and ^{39}Cl were found on the Al foil. This could only have come from the LAr bath. Since there is no way to be determine how much of the excess ^{24}Na cross section measured in the Al foil is from contamination (which would not affect the cross section determinations from the LAr cell) and how much is from secondary particles (which would affect the cross section determinations), the observed cross sections for the isotopes produced from the argon may only be interpreted as maximum values.

The Potassium Chloride monitor pellet: A pellet of KCl was mounted outside the apparatus in the beam, on the downstream side, for the purpose of additional monitoring of the reactions. KCl was chosen because the elements K and Cl flank Ar in the periodic table. Both the K and Cl are of smaller atomic weight than Ar, so neither ^{38}Cl nor ^{39}Cl could be produced from proton collision, and, indeed, no detectable quantities of those isotopes were present in the KCl pellet after the exposure. The only isotopes detected in the pellet were ^{24}Na and ^{28}Mg, with apparent cross sections 4.85(9) mb and 0.38(1) mb (average per nucleus).

Estimated discharge fractions: The fraction of an isotope which vented out of the cylinder with the argon gas is

$$F(\text{gas}) = \frac{\sigma(\text{Bubbler water})}{\sigma(\text{Reconstructed sum})}. \tag{11}$$

The fraction of an isotope which vented out of the calorimeter with the LAr is

$$F(\text{LAr}) = \frac{\sigma(\text{Reconstructed sum}) - \sigma(\text{Calorimeter rinse})}{\sigma(\text{Reconstructed sum})}. \tag{12}$$

The fractions (11) and (12) may be assumed to be the same for ^{22}Na and ^{24}Na, although the activity measurement in the bubbler water only produced an upper limit for ^{22}Na, resulting in the entry "< 13"(%) in Table 2 for F(gas). Conversely, the measurement of ^{22}Na yields a more precise estimate of the fraction discharged with the liquid, because it does not involve a reconstructed sum, and the value 44.6(9.9) may be taken for the ^{24}Na, which is more precise than the value 45(23) shown in the table.

Evidently, about half of the sodium vented out from the cell, through the plumbing, and mostly into the cylinder, while almost none vented out of the cylinder into the bubbler. It may be the mechanical venting process rather than the chemical properities of the isotopes which determined how much activity vented out of the cell and the cylinder. The argon in the cell was fairly rapidly cryopumped into the cylinder, while mostly still in a liquid state, while the argon in the cylinder was fairly slowly vented into the bubbler, completely in the gaseous state. The mechanical carrying or flushing capabilities of rapidly flowing liquid argon greatly exceeds that from gently venting gaseous argon. If the isotopes had fully attached to surfaces in the cell, this would not have mattered.

Under the assumption that all of the isotopes behaved similarly with respect to the two ventings, i.e., liquid from the cell and gas from the cylinder, the reconstructed cross sections for ^{28}Mg, ^{38}S, ^{38}Cl, and ^{39}Cl were formed by multiplying the contribution from the calorimeter rinse by the same ratio (9) used to reconstruct the ^{22}Na sum (10).

6 Conclusion

1. In case of an accident involving the release of argon gas from a calorimeter, less than 1% of the activity would be released from the calorimeter. More than 99% of the activity in a LAr calorimeter does not evaporate with the argon gas.

2. In case of an accident involving the release of LAr from a calorimeter, the bulk of the activity (well over 50%) would remain in the calorimeter.

    a. The activity we observed seems to have largely not attached to the calorimeter surfaces. This may be at least partly due to the short time the LAr sat in the cell during and following the exposure.

    b. Our test cell represents a worst-case as far as release of activity is concerned: it resembles a funnel, and released nearly half of its activity.

    c. A real calorimeter is full of crevices and surfaces, unlike our small test cell, and would trap more activity than our test cell.

   d. A real calorimeter will have its activity build up over a long period of time, rather than the hour or so over which our test cell was exposed and waited to be vented. The activity in a real calorimeter would therefore have much more time to attach to surfaces.

3. A worst-case scenario, in which a catastrophic dumping of activated LAr occurs, would result in contamination of any surfaces in the experimental hall onto which the LAr actually spilled and remained until boiling off. More than 99% of the contamination should remain on any such surfaces after the argon evaporates.

   A small contamination occurance during this experiment pertains to this. During the exposure, the test cell was immersed in a bath of liquid argon. The bath became quite radioactive, and activity was apparently coming out of the bath along with the evaporating argon. The detected activity which escaped the bath appears to have been almost exclusively ^{38}Cl and ^{39}Cl, which have half-lives less than 1 hour. Some quantity of the undetected isotopes ^{37}Ar, ^{39}Ar, and ^{41}Ar must also have escaped. It got on the floor of the exposure cave and onto the clothes of those manipulating the system (the clothes were confiscated by Health Physics and the personnel were decontaminated – overall, the contamination levels were fairly small). Based on the distribution of the contamination, the activity evidently did not rise above the level of the bath (i.e., it did not gain altitude). This is consistent with its identification as chlorine isotopes, which are heavier than air. By the next day, the activity of the clothing and the floor of the cave had decayed to background levels. The bath-vessel itself remains activated 4 months after the experiment. While this is at least partly due to its steel construction, most of this long-lived activity likely came from isotopes produced in the argon, since they could be collected by a sample swipe. However, it is important to point out that none of this long-lived activity escaped the bath-vessel. Therefore, if activated LAr spills onto a surface, there will be very little radioactivity that would be carried into the air, but the area where the LAr spills may be contaminated with long-lived activity.

   In such a worst-case, to prevent any airborne contamination above the cavern, we would recommend:

   a. Rapidly evacuate all personnel.

   b. Discontinue ventilation.

7 Acknowledgements

Our thanks to the staff at BNL's AGS for helping us with the exposure, including I. H. Chiang, D. Lazarus and K. Yip, and in particular P. Bergh and M. McAvoy from the Health Physics group, and to S. Kane for helpful suggestions for the cryogenic construction and operations. We would also like to thank K. Sexton for constructing the cell and S. Kurczak for carrying out the sample preparations and gamma counting.

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